

MOLECULAR CONDUCTORS

In-Plane Uniaxial Stress/Strain Modification of the Fermi Surface of (BEDT-TTF)₂KHg(SCN)₄

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We are employing a new device¹ to study the in-plane effect of uniaxial stress/strain on the Fermi surface properties of molecular conductors. We present in this brief report an example of such effects on the material (BEDT-TTF)₂KHg(SCN)₄. This material has a combination of closed orbits and open orbits in its Fermi surface. The open orbit sheets run parallel to the c-axis of the crystal in the normal metallic state of the material. Below a transition temperature of about 8 K, the Fermi surface reconstructs, and such properties as the magnetoresistance and the quantum oscillations take on a highly peculiar character. Some of these features are seen in Figure 1 at zero stress. Here there is a very large magnetoresistance (MR) followed at higher temperatures by complex

Shubnikov - de Haas oscillations (SdH). Above about 22.5 T the character of both the MR and the SdH change dramatically. It is generally thought that for higher fields the low temperature low magnetic field ground state is broken at this point, although many questions remain.²

The scientific interest in applying stress in-plane to this material is to try to alter the anisotropic character of the open and closed orbit Fermi surface topology. We have recently predicted, based on strain dependent band structure calculations, the effects of in-plane stress for guidance.³ In particular, there appears to be an important interplay between the two bands (open vs. closed) where such mechanisms as magnetic breakdown behavior in the SdH effect and open orbit carrier reservoir effects in the quantum Hall behavior are observed. Hence the ability to alter the inter-band band width and/or k-space separation in a controllable manner will allow us to study more carefully these effects.

We present below some preliminary results of this investigation. The sample axes are first determined by polarized FTIR spectroscopy, and then mounted in the uniaxial stress apparatus⁴ with the lateral stress translator.¹

We note that the stress is increased incrementally during the experimental run without need for cycling to room temperature. The method involves sweeping the magnetic field for specific stress values at different temperatures to obtain the information necessary for Fermiology-based studies. In particular the frequency of the SdH oscillations gives information about the change of Fermi momentum of the closed orbits with stress, and indirectly about the change in the unit cell. The temperature dependence of the SdH amplitude yields information about the effective mass of the carriers, and the background magnetoresistance tells us how the ground state is changing with stress. Returning to Figure 1, we see the effects of stress on the magnetotransport properties of the title material, for three values of stress applied along the c-axis. The systematic removal of the zero stress ground state signatures in the magnetotransport is quite clear. By 7 kbar we believe the ambient pressure-reconstructed ground state is completely removed.

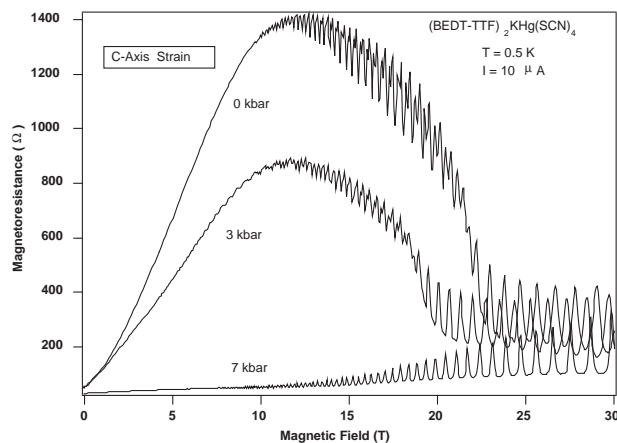


Figure 1. Magnetoresistance of the title material at low temperatures for three different values of uniaxial stress/strain applied along the c-axis. Details are discussed in the text.

In Figure 2 we show some detail of the high field behavior of the SdH oscillations. One can clearly see the change in the frequency, which is related to the area of the closed orbits in k-space (which increase in agreement with Reference 3). There is clearly one other dramatic effect—namely that the high stress data shows the lowest background magnetoresistance along with very flat minima and

very peaked maxima in the quantum oscillation wave form. It is generally believed that it is the influence of the open orbit band which degrades the approach to quantum Hall-like behavior in the SdH oscillations, and which contributes to a finite background magnetoresistance in the otherwise dissipationless regions in the middle of a particular Landau level. Here we see that such effects are reduced at high c-axis stress, and one possible reason is that this has reduced either the open orbit density of states, and/or the ability of the open and closed orbits to communicate across the momentum gap on the Fermi surface. Experimental work to investigate a-axis stress, and computational work to provide a more systematic description of the observed effects is in progress.

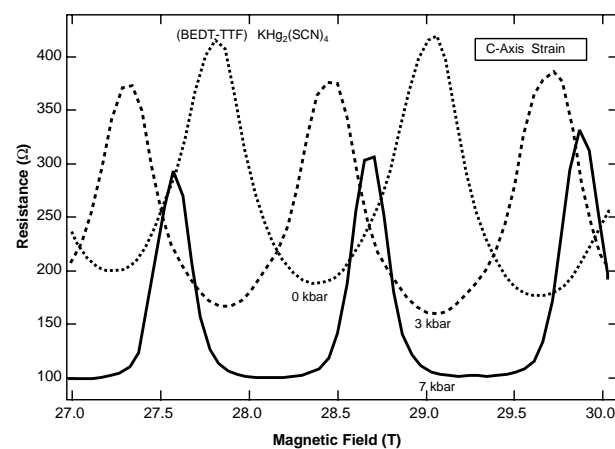


Figure 2. Magnified detail of the high field behavior of the SdH oscillations from Figure 1. Of note is the shift in the Landau levels with strain and the unusually flat minima in the 7 kbar data in each Landau level. Note further that the amplitude of the wave forms does not degrade, which is an indication that the strain is very uniform within the single crystal.

This work is supported by NSF-DMR 95-10427.

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“Kink Temperature” Characterization of the Phase Diagram of (BEDT- TTF)₂KHg(SCN)₄

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The purpose of this work has been to investigate the mechanism whereby either high magnetic field or temperature may remove the highly unusual ground state of the title material.¹ Briefly, the ground state may be summarized thusly. Below about 8 K an anomaly appears in the otherwise metallic the metallic behavior of the resistance, as shown in Figure 1 for the zero magnetic field case. This is interpreted as the nesting of part of the Fermi surface of this material, and a subsequent reconstruction of the remaining Fermi surface. Magnetotransport below 8 K, as depicted in Figure 2 at low temperatures, shows that the Fermi surface takes on a complicated topology, and the observed quantum oscillations bears this out. There have been extensive studies to investigate this low

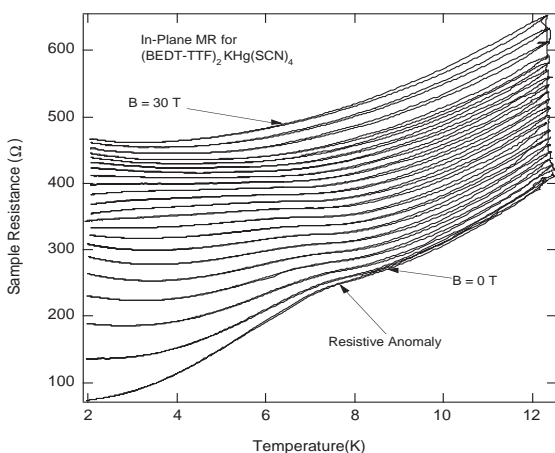


Figure 1. Resistance vs. temperature for (BEDT-TTF)₂KHg(SCN)₄ with magnetic field parallel to the conducting layers. Fields are from 0 to 30 T, and the resistive anomaly is shown as a bump in the temperature dependent resistance that starts at 8 K for zero field, and then moves to lower temperatures with increasing field.

temperature state,² but many uncertainties still remain about its exact nature.³ We note that in Figure 2 the magnetic field is *perpendicular* to the conducting layers of this highly anisotropic molecular conductor, and that at around 22.5 T there appears an anomaly in the magnetoresistance known as the “kink field” that has generally been interpreted as the removal of the anomalous ground state by the magnetic field.

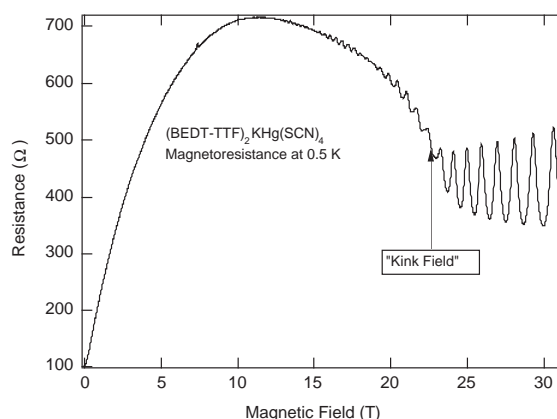


Figure 2. Representative magnetoresistance of the title material showing the quantum oscillations and the behavior of the “kink field” feature. The latter has been interpreted as the breaking of the low temperature, low magnetic field ground state by the magnetic field. The magnetic field is perpendicular to the conducting layers in this measurement.

We have undertaken systematic measurements of the temperature dependent resistivity of this material with the magnetic field *parallel* to the conducting layers over a range of field well in excess of the kink field. Such a study has two important considerations. The first is simply that in this orientation the magnetoresistance, which becomes large below 8 K as Figure 2 shows, is greatly reduced. This allows us to investigate the temperature anomaly “kink temperature” without the additional magnetoresistance signal. And second, such a study gives information on the isotropic (Zeeman) vs. anisotropic (orbital) nature of the mechanism that destroys the anomalous ground state. Since this is a quasi-two dimensional metal with open and closed orbit bands and with very little interplane band width, the phase line should be highly anisotropic in field direction if it is orbital in origin.

Our experimental data are shown in Figure 1 for the in-plane resistance vs. temperature over a range of magnetic fields that extend well above the “kink field.” Similar results were obtained for a number of different orientations of the magnetic field with respect to the in-plane axes. A summary of the main features of the data is shown in Figure 3, compared with a similar study for the field perpendicular to the conducting planes,¹ and with “kink field” magnetoresistance data also in the perpendicular configuration as in Figure 2. The points in Figure 3 were extracted from the data in Figure 1 by taking the derivatives of the curves to determine the position of the resistive anomaly more exactly for each field value. The main results of this study are

- that the phase transition line, as determined by the resistive anomaly (“kink temperature”), is isotropic in field direction, and
- that the “kink temperature” phase line extends beyond the point of the “kink field” at the lowest temperatures.

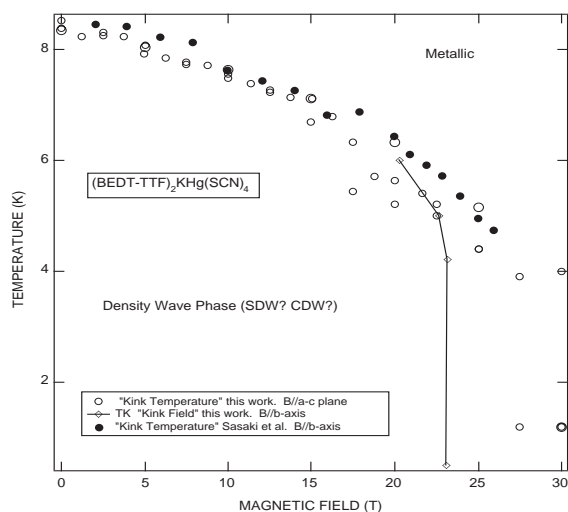


Figure 3. T-B phase diagram from our results. The open circles represent the “kink temperature” feature for three different directions of the magnetic field in-plane. The closed circles are similar data, but for the field perpendicular to the conducting planes by Sasaki *et al.* The solid line is the “kink field” line as determined from measurements as shown in Figure 2.

These measurements therefore indicate that the anomalous ground state, as determined from the kink temperature data, may extend well above 22.5

T, perhaps up to 30 T or more. Hence (Phe kink field anomaly may not be an accurate indicator of the destruction of the ground state. The isotropic nature of the kink temperature phase line supports a Zeeman-type mechanism, rather than a orbital (or nesting type) mechanism to destroy the anomalous ground state. Given the recent very complex descriptions of the phase diagram of this material from magnetization and transport studies,¹ high field NMR data,⁴ and reports of the quantum Hall effect above the kink field,⁵ the nature of the ground state in this material is still a subject of great controversy.

This work is supported by NSF-DMR-10427.

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High-Field NMR Measurements of the Spin-Peierls Phase of (TMTTF)₂PF₆

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Even though (TMTTF)₂PF₆ is isostructural to the Bechgaard salts (TMTSF)₂X, its conductivity is comparatively very small with an activated temperature dependence as high as 300 K. Evidence for a spin gap does not appear until temperatures below about 17 K, indicating the

onset of a spin-Peierls phase.¹ We report on the high-field properties of this material, as observed using ^{13}C NMR spectroscopy recorded at the solid state NMR facility at the NHMFL in Tallahassee.² Relaxation rate data were collected at the NHMFL Pulsed Field Facility in Los Alamos. The samples were ^{13}C -labeled on the bridging carbons of the dimer molecule.

In Figure 1, we show four spectra that demonstrate some of the effects of the spin-Peierls transition on the NMR spectrum. They are recorded under different conditions, so each is discussed individually. The top trace (a) is a spectrum recorded at $T = 35\text{ K}$ and 21.5 T . Two distinct lines are seen, corresponding to the inequivalent paramagnetic shifts of the two ^{13}C nuclei in each molecule.

The second spectrum (b), showing a single line, was taken at 16.5 T . The change from 35 K is a result of the phase transition and the associated vanishing (negative) paramagnetic shift. Finally, the lowest two traces ((c) and (d)) are broad spectra recorded at $T = 5\text{ K}$ and 12 K , respectively with the field centered near $B = 21.5\text{ T}$. The data were obtained by time-integration of the real part of the

spin echo and stepping the field, which became necessary because the linewidths were broad compared to the pulse bandwidths. The broadening was seen above a critical field $B_c \sim 19.1\text{ T}$. The spectra are consistent with an incommensurate phase with a linewidth of order 500 kHz – 1 MHz , corresponding to an internal field *variation* of about 1000 gauss at the ^{13}C nuclei for $T = 5\text{ K}$. The incommensurability is believed to result from the formation of domain walls consisting of a triplet excitation in each stack of TMTTF molecules. The onset of the strong temperature dependence to the linewidth appears to be at 12.5 – 13 K for $B > 20\text{ T}$, which we identify as the transition temperature to the incommensurate phase.

The work at UCLA was supported in part by NSF grants DMR 9412612 (SB), DMR 9319304 (WGC), and INT-9421019.

References:

- 1 See, for example, Creuzet, F., *et al.*, *Synth. Metals*, **19**, 289 (1987), and ref. therein.
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NMR Investigation of the Critical Fluctuations of the Spin-Density-Wave Transition in $(\text{TMTSF})_2\text{PF}_6$ at High Magnetic Field

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One of the hallmarks of the physical situation that determines a phase transition is the exponent that characterizes the temperature dependence of the divergence of the order parameter in the critical region close to the transition. In this report we describe preliminary results for the critical fluctuations associated with the spin-density-wave (SDW) transition in the organic conductor

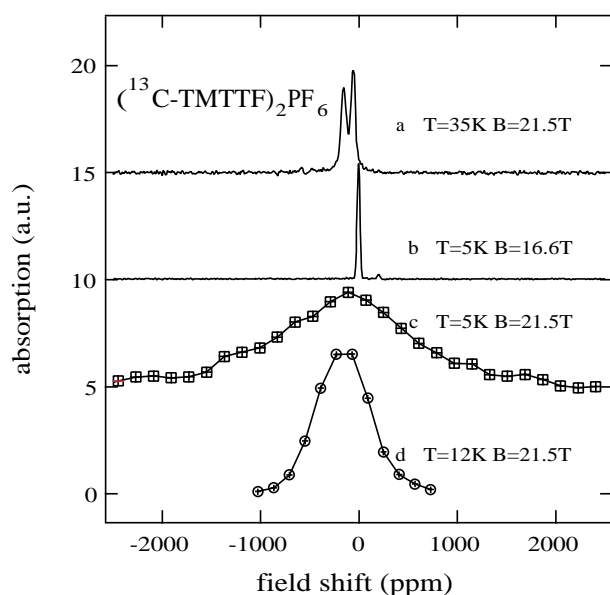


Figure 1. ^{13}C spectra in $(\text{TMTTF})_2\text{PF}_6$ taken at different fields and temperatures. The relative shift from zero in parts per million arises from the hyperfine coupling.

(TMTSF)₂PF₆ at high magnetic fields for temperatures (T) just above the SDW transition temperature ($T_s = 12.5$ K). The quantity measured is the proton spin-lattice relaxation rate ($1/T_1$) as a function of the temperature (T). The main results are that the critical fluctuation contribution to $1/T_1$ depends rather weakly upon the magnetic field alignment and shows a divergence in the vicinity of the transition, as reported earlier for substantially lower values of the magnetic field.¹⁻³ This divergence is shown in Figure 1, where $1/T_1$ is plotted as a function of $T - T_s$.

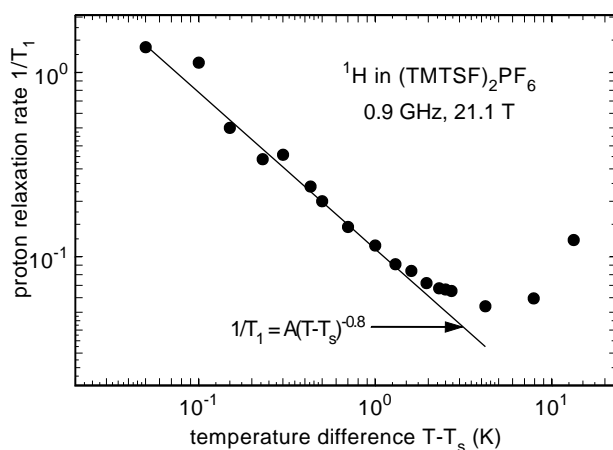


Figure 1. Proton spin-lattice relaxation rate as a function of T in the critical regime near T_s .

The solid line shows a divergence whose exponent is -0.8 that provides a reasonable fit to the data in the critical regime. This value is substantially different from the value -0.5 reported earlier based upon measurements at much lower fields.¹⁻³ It should also be pointed out that the magnitude of this critical component of $1/T_1$ is about the same for all frequencies investigated.⁴

These observations suggest that the critical order parameter fluctuation that couples to $1/T_1$ is primarily the amplitude and that its correlation time is shorter than the shortest time scale of our measurements ($\approx 2 \times 10^{-10}$ s). Such behavior is very different from that observed in the ordered SDW phase, where the coupling is to phase fluctuations that exhibit a very wide range of correlation times.^{4,5}

At present, it is not clear why the critical exponent at high field differs so much from that reported at

low magnetic fields. This issue is the subject of continuing investigations that, we hope, will clarify the effect of the magnetic field on the properties of the transition.

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Electron-Electron Interactions in Q1D Organic Conductors

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This is a continuation of the studies in Reference 1. Their purpose is to draw a conclusion whether or not in Q1D organic conductors interactions are so strong that traditional approaches to their thermodynamic and kinetic characteristics, which treat the materials as strongly anisotropic but common metals, could fail. Numerous speculations look for explanations of some unexpected transport and other properties of the Bechgaard salts in terms of "Luttinger Liquid."

As explained in Reference 1, any analysis of experimental data is to take into account some specific facts originating from rather strong anisotropy of the band structure and other Q1D features of electronic spectrum, characteristic of these materials.

In Reference 2, an excellent fit for the recently measured experimental temperature dependence of the longitudinal (i.e., along the chains' direction) resistivity of (TMTSF)₂PF₆ in the metallic phase under pressure, was obtained by taking into consideration the nesting features of

the Fermi surface in these materials. Experimental data display considerable deviations from the Fermi liquid T^2 dependence. The strength of electron-electron interactions, as evaluated from the fits, is rather small. The results do not support the idea of strong correlations, at least, in the TMTSF-salts.

Anomalous frequency dependence in conductivity is predicted for some Bechgaard salts in Reference 3. Numerical calculations are planned for frequency dependence of conductivity at finite temperatures.

References:

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Prediction of Effective Mass in Quasi-Two Dimensional Organic Conductors with Magnetic Breakdown

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The magnetic breakdown (MB) in low dimensional organic metals has been attracting much interest recently. Even though a lot of experiments and numerical calculations have been done about it, the origin of this effect has not been solved yet. In this report we try to give a proper method to investigate this problem and with its result we also numerically predict the effective mass of the material.

α -(BEDT-TTF)₂KHg(SCN)₄ and κ -(BEDT-TTF)₂Cu(NCS)₂ are the most popular materials in the BEDT-TTF salts (ET) family which have simple low dimensional Fermi surfaces (FS). Their FS contain 2D closed orbit and 1D open sheet.

The de Haas-van Alphen (dHvA) effect measurements of these materials show MB effect and the existence of anomalous oscillation frequencies that cannot be explained by existing theories. There are several attempts to solve this problem by using semiclassical theory or by simplifying it as a one band problem with quantum treatment, but they don't seem to give us satisfying results.

We construct a tight-binding Hamiltonian based on realistic crystal structure. The present model provides a natural description of magnetic breakdown phenomenon between co-existing closed and open Fermi surfaces in these materials. We compute the field dependence of magnetization and show that the dHvA frequencies predicted by our model in these conductors also account for the experimentally observed frequencies that are forbidden in the semiclassical picture. We argue that these semiclassically forbidden frequencies have a quantum mechanical origin and arise from the field dependent interplay between two bands that cross the Fermi energy. We extend the calculation to include the effect of finite temperatures on the field dependent magnetization and compute the value of effective mass for these organic conductors.

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De Haas-van Alphen Effect in the Organic Metal κ -(BEDT-TSF)₂Cu[N(CN)₂]Br

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De Haas-van Alphen experiments in pulsed magnetic fields are valuable for finding out new information about the electronic structure of organic metals. κ -(BEDT-TSF)₂Cu[N(CN)₂]Br is

a particularly interesting example, since it is a virtual structural analogue of the highest T_c BEDT-TTF-based superconductor.^{1,2} Somewhat unusually, the BEDT-TSF salt does not superconduct at all, in spite of the fact that its Fermi-surface is expected to be nearly the same.

Figure 1a shows an example of de Haas-van Alphen oscillations measured in fields up to 50 T. The node in the oscillations at ~ 38.5 T corresponds to cancellation of the frequency contributions from the “neck” and “belly” of the warped quasi-two-dimensional Fermi surface. At this field the bandwidth W of the Landau tubes is approximately equal to half of the cyclotron energy. Below this field the pattern of quantum oscillations consists of beats (Figure 1b), whereas above this field the system behaves as a two-dimensional metal (Figure 1c).

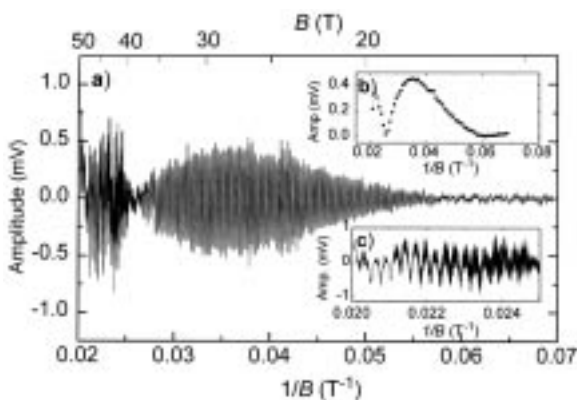


Figure 1. (a) An example of de Haas-van Alphen oscillations measured in κ -(BEDT-TSF)₂Cu[N(CN)₂]Br in pulsed magnetic fields. (b) The amplitude envelope of the oscillations, revealing the beat structure. (c) Cusp-shaped oscillations in the susceptibility at the highest fields characteristic of a two-dimensional metal.

Studies of the temperature dependence of the quantum oscillations reveal that the effective mass is much smaller than in the equivalent BEDT-TTF salt. This likely reflects the differences in electron-electron and electron-phonon interactions between the two salts. The persistence of the oscillations to lower fields, however, implies that the BEDT-TSF salt is of higher purity. Difficulties with the preparation

of κ -(BEDT-TTF)₂Cu[N(CN)₂]Br single crystals have been a major obstacle preventing detailed Fermi-surface studies.

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Fermiology of Organic Conductors Containing Magnetic Ions

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The inclusion of magnetic ions adds a new dimension to the phase diagram of possible ground state properties of charge-transfer salts. κ -(BEDT-TSF)₂FeCl₄ is one such example of a magnetic charge-transfer salt which, in spite of the existence of antiferromagnetic interactions between the Fe moments, retains a metallic ground state at low temperatures.¹ The λ -(BEDT-TSF)₂FeCl₄ salt, on the other hand (note the different crystalline phase), undergoes a metal to insulator transition on cooling below 8 K.²

Whereas quantum oscillations have not been observed in λ -(BEDT-TSF)₂FeCl₄, they can be observed in κ -(BEDT-TSF)₂FeCl₄. The most noticeable effect of the magnetic ions in the latter salt is the enhancement of the effective mass by approximately a factor of 2. This can be inferred by comparing the temperature dependence of the quantum oscillations with those in the isostructural non-magnetic salt in which Fe has been replaced by Ga.³ Spin fluctuations involving the Fe moments is the likely reason for the mass enhancement. Figure 1 shows a typical example of

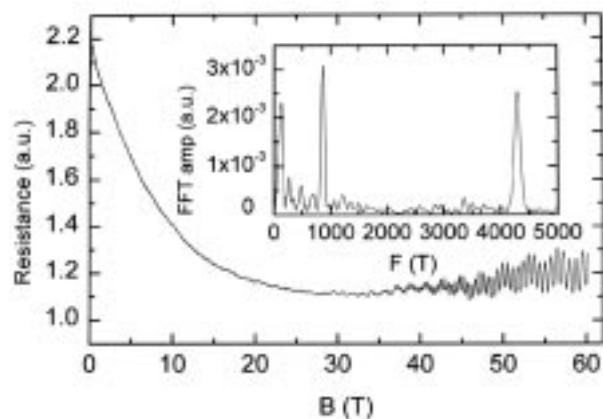


Figure 1. An example of the magnetoresistance of κ -(BEDT-TSF) $_2$ FeCl $_4$ in pulsed magnetic fields, with the Fourier transform shown in the inset.

Shubnikov-de Haas oscillations measured at 1.5 K in fields of up to 60 T. Note the pronounced negative magnetoresistance at low magnetic fields, indicative of a magnetic scattering mechanism.

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κ -(BEDT-TTF) $_2$ I $_3$: A Quasi-Two-Dimensional Spin-Split Fermi Liquid in Strong Magnetic Fields

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κ -(BEDT-TTF) $_2$ I $_3$ is one of the most two-dimensional of all organic conductors, with many of the crystals possessing exceptionally long mean

free paths. These facts make κ -(BEDT-TTF) $_2$ I $_3$ particularly interesting for the study of effects associated with two-dimensional electron systems in strong magnetic fields. Indeed, the Shubnikov-de Haas oscillations have already been observed to behave in an anomalous fashion, and previous publications have even suggested the existence of quasiparticles with fractional statistics.¹

Magnetization measurements in Los Alamos yielded very large oscillations, with a pronounced eddy current contribution in addition to the de Haas-van Alphen response. While the presence of eddy current resonances indeed suggest a possible connection with the quantum Hall effect, which is not too distantly related to the concept of quasiparticles with fractional statistics, the de Haas-van Alphen oscillations are behaviorally characteristic of a two-dimensional spin-split Fermi-liquid. Figure 1 shows an example of the de Haas-van Alphen component extracted from the measurement. When the Landau levels are not spin-degenerate, the oscillatory chemical potential has the effect of causing the spectral weight to be shifted from the fundamental onto the harmonics, leading to an apparent suppression of the effective mass at low temperatures and strong magnetic fields.

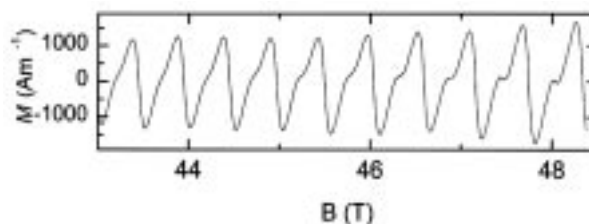


Figure 1. An example of spin-split saw-tooth de Haas-van Alphen magnetization oscillations measured in κ -(BEDT-TTF) $_2$ I $_3$ in pulsed magnetic fields at ~400 mK.

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Quantum Hall Effect in BEDT-TTF Salts

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A number of recent papers have suggested the existence of the quantum Hall effect in α -(BEDT-TTF)₂MHg(SCN)₄ salts, where M = K or Tl.^{1,2,3} This was inferred from the observation of quasipersistent currents in the magnetization¹ and the unusual behavior of the longitudinal magnetotransport in the high field phase.³ No direct observation of quantized Hall plateaux had yet been made.

The water-cooled Bitter magnets at Tallahassee provided the best conditions in which to attempt to observe the quantum Hall effect. The static fields of up to 33 T were sufficient to drive the materials under investigation well into the high field state. At the same time, the polarity of the magnetic field could be reversed, enabling the Hall component to be extracted by subtracting negative and positive field sweeps. In addition to the crystal that had previously yielded the quasipersistent currents,¹ transparently thin crystals were also used for the investigation, so as to maximize the Hall resistance.

Figure 1a shows raw resistance data of α -(BEDT-TTF)₂TlHg(SCN)₄ measured for both field polarities. We arrive at the in-plane resistivity and the Hall resistivity in Figure 1b upon subtraction and addition of the positive and negative field data. Note that the hysteresis, characteristic of this material, is clearly observable. The strong oscillations of the Hall resistance are a manifestation of the additional quasi-one-dimensional states, which play an important role in these materials.^{1,2}

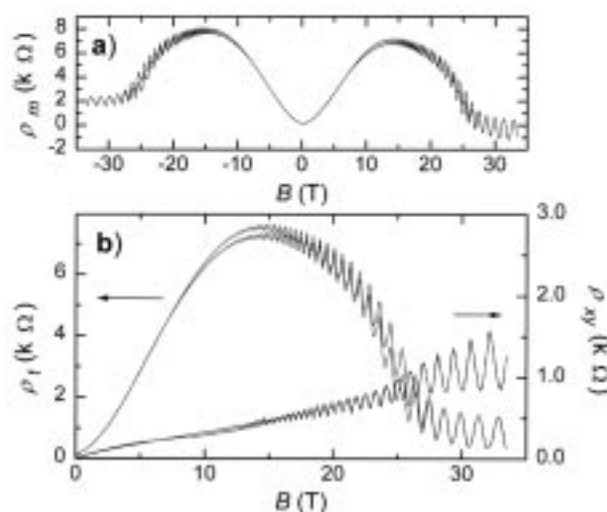


Figure 1. (a) The raw resistance data measured on α -(BEDT-TTF)₂TlHg(SCN)₄ at 0.7 K. (b) The in-plane and Hall resistivity components after taking account of the polarity of the magnetic field.

Only when we performed measurements on the exceptionally high quality sample in which the quasipersistent currents were originally observed, were plateaux observed, and only at fields above 30 T.⁴ Because of the quasi-one-dimensional states,² deep minima exist between the plateaux in Figures 2a and 2b. As a general requirement of the quantum Hall effect, the plateaux coincide with the minima in the in-plane resistivity (Figure 2c), which themselves coincide with the induced currents (Figure 2d) in the magnetization.¹ The

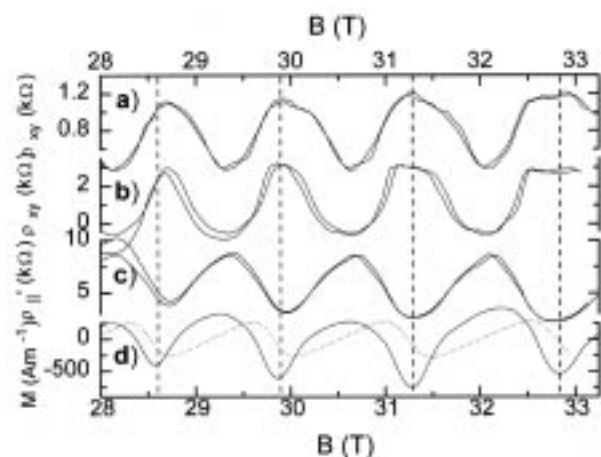


Figure 2. (a) and (b) The Hall resistance in another sample for two different contact configurations, indicating plateaux. (c) An example of the in-plane resistivity. (d) The corresponding magnetization measured in pulsed magnetic fields.¹

dotted line in Figure 2d illustrates the expected magnetization in the absence of the induced currents.

References:

- 1 Harrison, N., *et al.*, Phys. Rev. Lett., 77, 1576 (1996).
- 2 Harrison, N., *et al.*, J. Phys. Condens. Matter, 9, L47 (1997).
- 3 Hill, S., *et al.*, Phys. Rev. B, 55, R4891 (1997).
- 4 Harrison, N., *et al.*, Phys. Rev. B, 55, R16005 (1997).

Electrodynamic Studies of Unconventional Molecular Conductors

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We have used a cavity perturbation technique to explore various low-temperature phases of a series of organic charge transfer salts. Experiments essentially measure the complex conductivities of small single crystals (see *1996 NHMFL Annual Report*), over the frequency range from about 25 to 110 GHz. We have found that this technique is especially suited to highly anisotropic conductors in that it reliably enables independent measurements to be made for each diagonal component of the conductivity tensor; this is extremely difficult to achieve by DC methods, where finite size (comparable to sample dimensions) electrical contacts must be made to the sample. Furthermore, we are able to probe the frequency dependence of the complex conductivity by these methods.

Measurements in the millimeter-wave spectral range are particularly pertinent to organic conductors since the corresponding frequencies are comparable to typical electronic relaxation rates at liquid He temperatures.¹ Recent studies have shown that the low-temperature conducting phases in many organic conductors [*e.g.* the

α -(ET)₂MHg(SCN)₄ and (TMTSF)₂X salts] may be highly unconventional.^{2,3} To test this, we have made comparisons between the DC and AC magneto-transport properties of several of these compounds. For a conventional metal, there is no reason to expect any qualitative differences in the AC and DC responses.

Figure 1 shows data obtained for α -(ET)₂TlHg(SCN)₄, at a frequency of ~47 GHz. The data represent possible phase boundaries, as determined by drastic changes in the AC magneto-transport properties, *e.g.* pronounced maxima, minima, or kinks in the AC magneto-conductivity. The squares in the upper panel agree with previous DC measurements, while the circles agree with one recent study.⁴ On the other hand, the phase boundary in the lower figure has not been reported previously (either DC or AC) and seems to suggest that the transport properties parallel, and perpendicular, to the two-dimensional conducting layers are rather different. At present, it is not clear why the AC measurement is able to resolve these new features, however, it is clear that this is indicative of highly unconventional behavior.

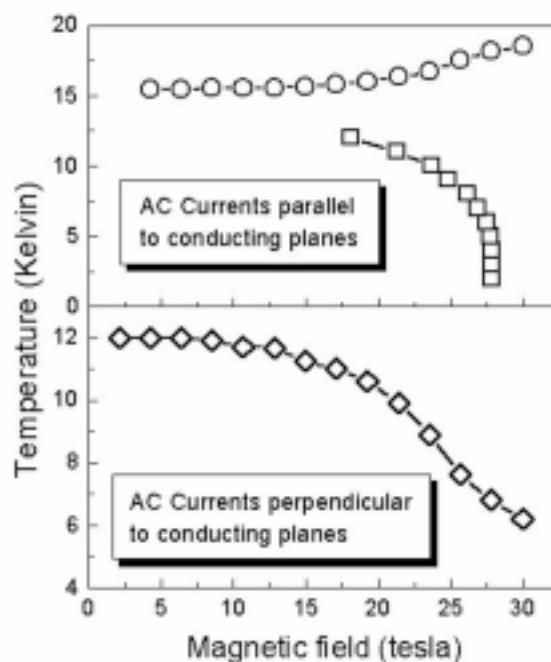


Figure 1. Data obtained for α -(ET)₂TlHg(SCN)₄, at a frequency of ~47 GHz.

References:

- 1 Hill, S., *et al.*, Proc. SPIE, **2842**, 296-306 (1996).
- 2 Danner, G.M., *et al.*, Phys. Rev. Lett. **75**, 4690 (1995).
- 3 McKenzie, R.H., *et al.*, cond-mat/9710245 (23rd October 1997).
- 4 Sasaki, T., *et al.*, Physica B, **216**, 366 (1996).

Angle-Dependent Magnetoresistance Oscillations in Organic Charge-Transfer Salts Exhibiting Field-Induced Phase Transitions

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This work continues a three-year program in which angle-dependent magnetoresistance oscillations (AMRO) are used to characterize the magnetic-field-induced transitions in a variety of organic charge-transfer salts. In contrast to magnetic-quantum oscillations (e.g. de Haas-van Alphen), which only give the areas of possible closed semiclassical orbits about the Fermi surface, AMRO enable both closed and open sections of Fermi surface to be observed,¹⁻³ and show in a straightforward manner whether the Fermi surface is being reconstructed at a transition.

Much of the work carried out by this collaboration at Tallahassee in 1996 and the first part of 1997 has involved AMRO¹⁻³ and magnetoresistance⁴ measurements of α -(BEDT-TTF)₂MHg(SCN)₄ (M=K, Tl). These salts have been the subject of

much controversial discussion concerning the nature of the low-field, low-temperature groundstate and the transition to a high field state at the so-called kink field. AMRO experiments carried out at Tallahassee in 1996-97 have been invaluable in characterizing the Fermi surface on either side of the kink, and a schematic picture is being built up that can explain most of the data obtained.¹⁻⁵ Some of the latest developments include an extension of the technique to GHz frequencies back in Oxford, leading to the discovery of a new type of magnetic resonance, the Fermi-surface Traversal Resonance (FTR).⁵ The semiclassical formalism developed to explain AMRO⁶ can also successfully predict the FTRs,⁵ demonstrating that the Fermi surface of α -(BEDT-TTF)₂KHg(SCN)₄ can be understood without recourse to exotic mechanisms such as incoherent interlayer tunnelling.

In the latter part of 1997, attention has turned to the two-dimensional superconductor (BEDO-TTF)₂ReO₄H₂O, which appears to undergo some form of Fermi surface reconstruction at fields of around 18 T.⁷ A large quantity of magnetoresistance data have been acquired in the 20 MW magnets at Tallahassee using a cryostat built in Oxford that allows the sample to be rotated through all possible orientations in the field. The data are at being prepared for publication.⁷

References:

- 1 Hayes, W., *et al.*, Synthetic Metals, **86**, 1949 (1997).
- 2 Honold, M.M., *et al.*, Synthetic Metals, **86**, 2055 (1997).
- 3 House, A.A., *et al.*, J. Phys.: Condensed Matter, **8**, 8829 (1996).
- 4 Harrison, N., *et al.*, Phys. Rev. B, **55**, R16005 (1997).
- 5 Ardavan, A., *et al.*, Physical Review Letters, submitted.
- 6 Blundell, S.J *et al.*, Journal de Physique I, **6**, 1837 (1996).
- 7 Audouard, A., *et al.*, Journal de Physique I, in press, and to be submitted.

Observation of the Beta-Frequency in the High-Field State of α -(BEDT-TTF) $_2$ KHg(SCN) $_4$

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The low-temperature ground state of the organic charge-transfer salt α -(BEDT-TF) $_2$ KHg(SCN) $_4$ has been the subject of much controversial discussion. It is now widely believed that a spin-density wave (SDW) state is formed resulting from nesting of the one-dimensional sheets of the high-temperature Fermi surface.¹ On the application of magnetic fields in excess of $B \approx 24$ T, the salt reverts back to a state that is experimentally indistinguishable from the high-temperature phase.¹

One of the main difficulties of this model, posed by experiment, has been the observation of a frequency of 4250 T, usually referred to as the β -frequency [see for example, References 1,2]. This frequency has been seen in both magnetization and magnetotransport measurements and is hence associated with a real k -space orbit. The orbit appears to relate to a complete breakdown between the one- and two-dimensional parts of the unnested Fermi surface. However, all previous observations have been restricted to the SDW-state only, where the area of the nested Brillouin zone should be far too small to accommodate an orbit of this size.

Our recent magnetization measurements in the 60 T pulsed magnet system at NHMFL-LANL have for the first time revealed the existence of the β -frequency in the high-field state of α -(BEDT-TTF) $_2$ KHg(SCN) $_4$.³ In addition, combination

frequencies β - α and β + α have been observed (see Figure 1). This refutes the proposal that the β -frequency is a signature only of the low-field state. Indeed, in the measured sample, the β -frequency becomes much more prominent at higher fields in accordance with models of magnetic breakdown.³

Nonetheless, the observation of the β -frequency outside the SDW-regime is very difficult. One prerequisite is an exceptionally pure sample. The Fourier transform in Figure 1 has been derived from the average of 15 magnetization traces taken at a base temperature of 400 mK. These experimental difficulties may explain why no β -frequency has thus far been observed in the isostructural salts α -(BEDT-TTF) $_2$ MHg(SCN) $_4$, $M = \text{Tl}, \text{NH}_4$.

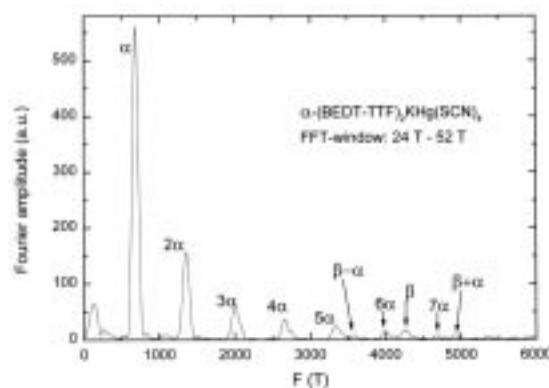


Figure 1. Fourier transform of the de Haas-van Alphen oscillations in α -(BEDT-TTF) $_2$ KHg(SCN) $_4$ in the field range of $24 \text{ T} < B < 52 \text{ T}$. The transformation has been carried out after averaging 15 shots taken at a temperature of 400 mK.

References:

- House, A.A. *et al.*, J. Phys.: Condens. Matter, **8**, 10361 (1996), *ibid.* **8**, 10377 (1996), *ibid.* **8**, 8829 (1996).
- Uji, S. *et al.*, Solid State Commun., **88**, 683 (1993).
- Honold, M.M., *et al.*, to be published

Pulsed Field Magnetotransport Measurements in α - $[(\text{CH}_3)_2(\text{C}_2\text{H}_5)_2\text{N}][\text{Ni}(\text{dmit})_2]_2$

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We have carried out measurements of the interplane magnetoresistivity in the quasi-two dimensional molecular metal α - $[(\text{CH}_3)_2(\text{C}_2\text{H}_5)_2\text{N}][\text{Ni}(\text{dmit})_2]_2$. Using the pulsed magnet systems at NHMFL-LANL, fields of up to 60 T were applied.

Fermiological studies of α - $[(\text{CH}_3)_2(\text{C}_2\text{H}_5)_2\text{N}][\text{Ni}(\text{dmit})_2]_2$ are problematic owing to a structural transition at 230 K, below which the Fermi surface changes shape. Previous band structure calculations¹ have suggested a highly complex low-temperature Fermi surface, which is bound to exhibit a multitude of Shubnikov-de Haas and de Haas-van Alphen frequencies. A combination of various magnetic breakdown frequencies is expected to govern the resistivity at the highest fields.

However, even at fields of up to 60 T, we do not find any new fundamental frequencies.³ The two main frequencies $\beta = 213$ T and $\gamma = 4081$ T appear to originate from a coupled magnetic breakdown network and dominate the oscillatory spectrum up to the highest fields (see Figures 1 and 2). We do not observe the $\delta = 520$ T frequency reported in Reference 2. Previously,^{1,2} the observed magnetic breakdown had been thought to occur between

quasi one-dimensional sheets and a closed hole pocket ascribed to the 520 T frequency.

This evidence suggests that the proposed low-temperature Fermi surface is not able to account for the observed magnetoresistive behavior. In fact, the actual Fermi surface may be much simpler than assumed, only comprising a single closed pocket and two quasi one-dimensional sheets.

The effective electron mass of the γ -orbit is derived to be $m_\gamma = 3.70 m_e$. The β -frequency, however, exhibits an almost constant Fourier amplitude over the measured temperature range ($0.4 \text{ K} < T < 6 \text{ K}$). Hence its effective mass is estimated to have an upper limit of $0.5 m_e$. Owing to this surprisingly low value, the possibility of the β -orbit being a Stark quantum interference orbit cannot be ruled out.

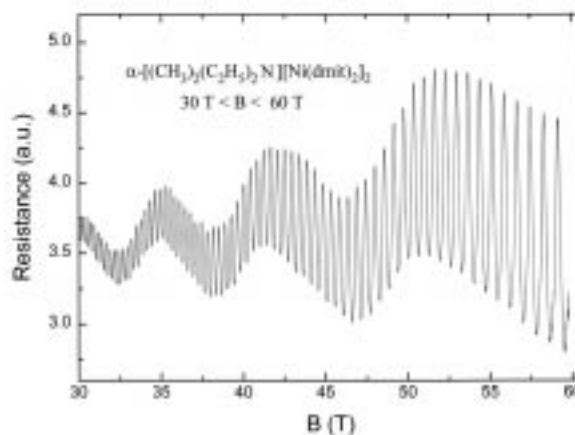


Figure 1. Interplane magnetotransport in α - $[(\text{CH}_3)_2(\text{C}_2\text{H}_5)_2\text{N}][\text{Ni}(\text{dmit})_2]_2$ at fields of $30 \text{ T} < B < 60 \text{ T}$ and $T = 500 \text{ mK}$.

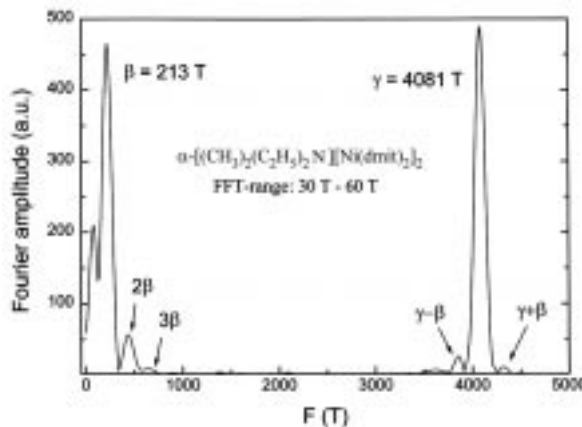


Figure 2. Fourier transform of the oscillatory component of the magnetoresistance for $30 \text{ T} < B < 60 \text{ T}$.

References:

- 1 Kobayashi, A., *et al.*, Phys. Rev. B, **51**, 3198 (1995).
- 2 Tajima, H., *et al.*, Solid State Commun., **88**, 605 (1993).
- 3 Honold, M.M., *et al.*, to be published.

Quantum Hall Effect and Chiral Fermi Liquids in Quasi-Two-Dimensional Organic Conductors at Very High Magnetic Fields

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The magnetization and magnetoresistance of single crystals of the quasi-two-dimensional organic conductor α -(BEDT-TTF)₂KHg(SCN)₄ have been measured at temperatures between 350 mK and 10 K in steady magnetic fields of up to 33 T and pulsed magnetic fields of up to 60 T. In its high field state (above ~25 T), this material has a Fermi surface consisting of a warped quasi-two-dimensional (Q2D) cylinder and a pair of quasi-one-dimensional (Q1D) electron sheets. The highest quality crystals exhibit a new form of Quantum Hall Effect (QHE), in which the chemical potential (Fermi level) is pinned to the Q1D states between very sharp Q2D Landau levels over finite regions of magnetic field. Above 30 T, the resistivity component ρ_{xy} consists of a series of Hall plateaux separated by minima caused by the influence of the Q1D carriers,^{1,2} in agreement with the predictions of calculations.¹ The QHE is also observed as a series of sharp negative spikes that occur in the pulsed-field magnetization data (see Reference 3 and references therein) whenever the chemical potential is between Landau levels. The negative spikes are due

to the deep minima in the in-plane resistivity, which allow quasi-persistent induced currents to flow.

The crystals in which the QHE occurs also show strong evidence for the presence of a chiral Fermi liquid at the sample edges.³ At low temperatures, the chiral Fermi liquid acts to short out the other current paths in the sample, causing a very dramatic attenuation of the peaks of the Shubnikov-de Haas oscillations in the resistivity component ρ_{zz} ; for example at 500 mK, the amplitude of the oscillations falls to around 10% of its maximum value (the maximum value is observed at 2 K). A careful analysis of the data allows the scaling behavior of the resistivity of the chiral Fermi liquid to be extracted. It appears that the resistivity scales as T^1 , suggesting a departure from conventional Fermi-liquid behavior. In contrast, the de Haas-van Alphen oscillations seem to be relatively unaffected by the presence of the edge states.³

References:

- 1 Harrison, N., *et al.*, J. Phys.: Condens. Matter, **9**, L46 (1997).
- 2 Harrison, N., *et al.*, Phys. Rev. B, **55**, R16005 (1997).
- 3 Honold, M.M., *et al.*, J. Phys.: Condens. Matter, **9**, L533 (1997).

New Aspects of the High Field Phase Diagram of α -(BEDT-TTF)₂KHg(SCN)₄ as Determined from ¹³C NMR

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α -(BEDT-TTF)₂KHg(SCN)₄ is a quasi-two-dimensional organic conductor that has attracted recent attention. While it remains metallic at low temperature, anomalies in the susceptibility,

magnetoresistance, and Hall coefficient are observed, and μ SR and other studies indicate the possibility of a phase boundary with a SDW with anomalously small moment, or a CDW, below 8 K. We have measured the ^{13}C spin lattice relaxation time, T_1 , in an enriched sample over the temperature range 3 K to 40 K, and at fields from 10 T to 23.25 T, crossing the phase boundary over a wide range of fields. The line shape of the central C's shows three inequivalent carbon sites in agreement with the low field work of Miyagawa *et al.*¹ at all fields and temperatures. No evidence of a SDW in the line shape is observed in the temperature and field range covered. This restricts the value of the moment to less than 0.03 BM, or the unlikely situation that the SDW are unpinned and moving rapidly, providing an averaging mechanism of the local field at the ^{13}C site. As shown in Figure 1, at 10 T and 15 T the T_1^{-1} behavior is Korringa like above 8 K with $(T_1 T)^{-1} = 8.2 \text{ (ms K)}^{-1}$, with a sharp drop at 8 K and non-Korringa behavior below 8 K. At 22.24 T, still below the published phase boundary, the same behavior is observed, but with $(T_1 T)^{-1} = 6 \text{ (ms K)}^{-1}$. At 23.25, above the phase boundary, $(T_1 T)^{-1}$ is 6 (ms K)^{-1} , but the evidence of the sudden decrease near 8 K is greatly reduced or absent. One can speculate that above the transition the Fermi surface is somehow reduced at high fields. Work is in progress to carry these measurements to 30 T, and to obtain line shift data to elucidate this interesting behavior.

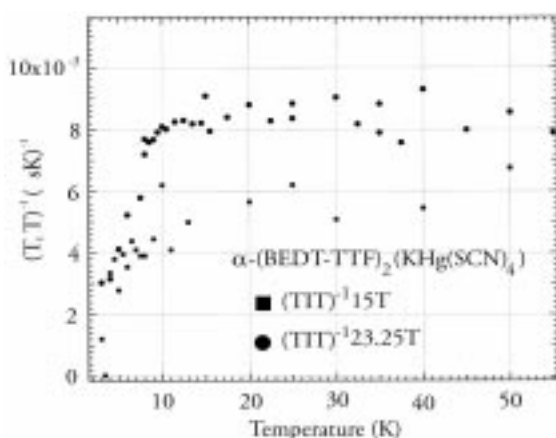


Figure 1. $(T_1 T)^{-1}$ of the α -(BEDT-TTF) $\text{KHg}(\text{SCN})_4$ as a function of temperature, at 15 T and 23.25 T.

References:

- 1 Miyagawa, K., *et al.*, Phys. Rev B, **56**, R8487 (1997).

Fermi Surface Measurements in the Organic Superconductor λ -(BETS) $_2$ GaCl $_4$

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The Fermi surface topology has been investigated in a unique organic superconductor λ -(BETS) $_2$ GaCl $_4$. Recently it has been discovered that λ -(BETS) $_2$ GaCl $_4$ has a non-s-wave pairing mechanism¹ of the superconducting quasi-particles. This study is interesting because we probe the nature of the Fermi surface and the metallic state of a superconductor with unconventional pairing (s or d-wave). We can address the question whether or not the normal metal state follows Fermi liquid behavior or not. The investigation of the Fermi surface (Shubnikov-de Haas effect (SdH)) was performed at the NHMFL-Los Alamos using rf transport techniques ($\sim 500 \text{ kHz}$) in a plastic ^3He refrigerator (340 mK base) in a 15 mm bore 60 T (8 ms) capacitively-driven coil.

Our results thus far indicate that Fermi liquid-like behavior is followed in the superconductor λ -(BETS) $_2$ GaCl $_4$. This determination stems from analysis of the temperature dependence of the SdH oscillations. The Fermi surface, however, shows significant deviations from calculated values. Figure 1 shows a closed orbit of 650 T and a breakdown orbit (which is equivalent to the area of the Brillouin zone (BZ)) of 4100 T. The theoretical values of areas of the orbit and the BZ zone, predicted from tight-binding band calculations based on an X-ray structure determined at 17 K,² were 829 T and 3768 T, respectively. These significant discrepancies will be addressed in further high field measurements at finite angles. Because the SdH oscillations first appear at fields near 40 T, pulsed fields are necessary for future investigations.

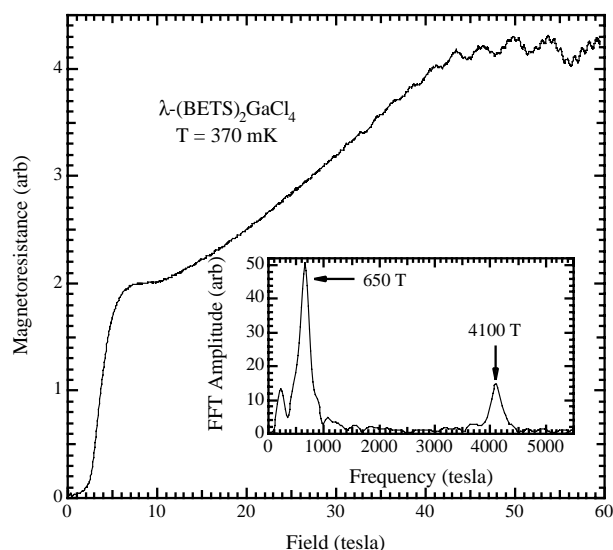


Figure 1. The magnetotransport of λ -(BETS) $_2$ GaCl $_4$ at 370 mK. The inset shows the FFT of the above waveform.

References:

- 1 Mielke, C.H., *et al.*, preprint (1997).
- 2 Kobayashi, A., *et al.*, unpublished results.

Fermi Surface Measurements of κ -(ET) $_2$ Cu[N(CN) $_2$]Br

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Transport measurements¹ performed at NHMFL-Los Alamos in fields of 60 T (see Figure 1) showed for the first time quantum oscillations in κ -(ET) $_2$ Cu[N(CN) $_2$]Br at ambient pressure. A careful analysis of the temperature dependence of the quantum oscillations indicates Fermi-liquid behavior. The observed frequency of 3798 ± 5 T corresponds to 100% of the Brillouin zone area (to within 0.1%). Low temperature crystallographic measurements and band structure calculations predict a closed hole pocket nested between two 1-D sheets. Magnetic breakdown between the closed pockets and the 1-D sheets result in an SdH frequency equal to the Brillouin zone. It is interesting to note that the measurements shown in

Figure 1 indicate a negative slope to the magnetoresistance. Further investigation in a DC magnet to 18 T showed a temperature dependence of the negative slope. In some model systems, a negative magnetoresistance is due to randomly oriented magnetic impurities. An applied magnetic field polarizes the impurities and reduces the scattering as the field intensity increases. Our observation is consistent with such behavior. It is well known that one of the starting materials in the synthesis of κ -(ET) $_2$ Cu[N(CN) $_2$]Br, namely CuBr, is susceptible to a decomposition process that results in conversion of Cu(I) to Cu(II). Cu(II), having a magnetic moment, is likely to be the source of the magnetic impurity. Further evidence supporting this hypothesis was obtained in magnetization and transport studies performed on crystals containing approximately 1% of Cu(II); these crystals showed no indication of the SdH effect. Magnetization measurements performed

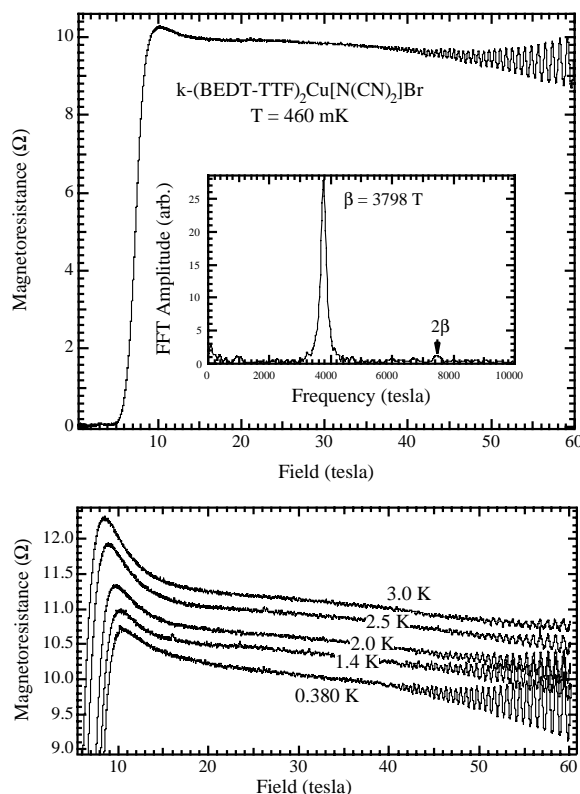


Figure 1. Magnetoresistance of κ -(ET) $_2$ Cu[N(CN) $_2$]Br, showing quantum oscillations beginning at ~ 38 T. Measurements were made at $T = 460$ mK. The inset shows the FFT amplitude with a peak at 3798 T. The lower figure shows the temperature dependence to fields of 60 T. Note the negative slope extending to the highest fields.

on crystals that showed the SdH effect indicated that Cu(II) concentrations were reduced to levels nearing the resolution of the SQUID magnetometer.

Determinations of the Dingle temperature and scattering length on crystals that exhibited quantum oscillations resulted in $T_D = 3.4$ K and a scattering length of 250 Å. This Dingle temperature is rather high and the scattering length is nearly an order of magnitude shorter compared to most organic superconductors which exhibit quantum oscillations. Magnetic impurities resulting in an increased Dingle temperature and a reduced scattering length easily could mask the SdH effect in earlier measurements and also give rise to the observed spin fluctuations.

References:

- ¹ Mielke, C.H., *et al.*, Phys. Rev. B, **56**, 4309 (1997).

Magnetotransport to 145 T of (TMTSF)₂ClO₄

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The magnetotransport of single crystal (TMTSF)₂ClO₄ has been measured in the quenched state to fields of 145 T at a

temperature of ~8 K. The magnetic field was created by use of a single stage flux compression, magnetic field generator.¹ The magnetic field generator, i.e. flux compression, is powered by approximately 4 kg of Detasheet™ C-4 plastic explosives and a 340 kJ capacitor bank. The capacitor bank is used to create the initial current pulse, while the explosives are used to compress the seed flux of the magnet into a smaller volume as well as crowbar the magnet circuit. The seed field risetime (0 T to 20 T) is approximately 50 μs in duration, while a 20 μs risetime occurs between 20 T and 145 T. The experiment terminates shortly after peak field when a jet of vaporized copper contacts the sample area at speeds of approximately 3000 m/s. The magnetotransport was measured by a DC technique that uses six parallel wires (3 for I and 3 for V) to perform a variation of a 4 lead ohmic measurement. The method exploits geometric symmetries which self compensate open loop areas, resulting in a maximum pick-up voltage of 400 mV (in this case). Such levels of pick-up are outstanding considering that the dB/dt induces more than 10 V per square mm at the sample. The sample was cooled by use of a plastic (PVC and G-10) liquid helium cryostat.

The magnetotransport results are shown in Figure 1. Despite the rather high temperature of 8 K, which was due to a vapor lock shortly before the shot, these data are the first measurements of this well known compound to such extremes of magnetic field. The data below approximately 35 T is unreliable due to the extreme electrical noise associated with the capacitor bank switching and the magnet crowbar. A digital signal processing algorithm was used to remove time space oscillatory signals resulting from transmission line reflections. A nearly linear magnetoresistance is observed between 70 T and 100 T; above 100 T a quadratic magnetoresistance is observed. Subtraction of a smooth background reveals weak quantum oscillations that agree with earlier results from 60 T non-destructive millisecond pulsed experiments.² The quantum (rapid)

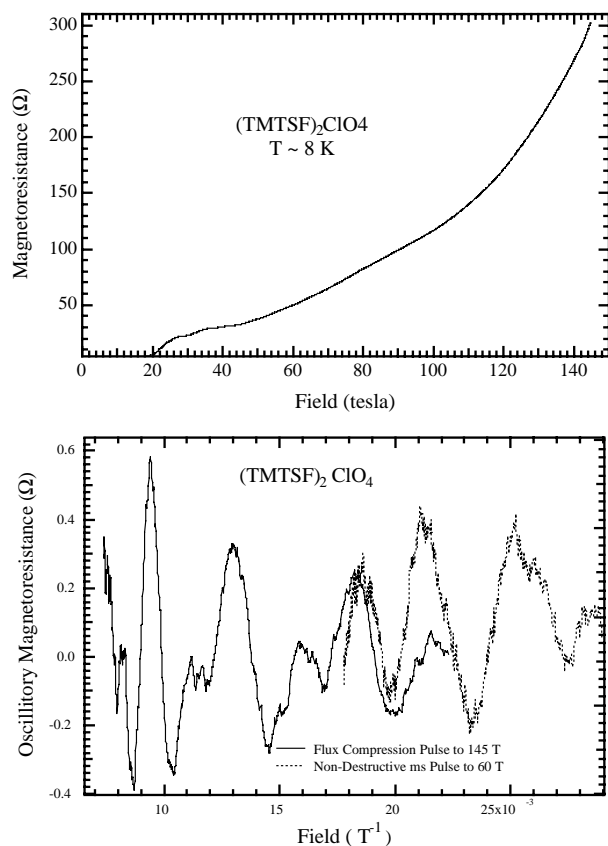


Figure 1. (Top) Magnetotransport of $(\text{TMTSF})_2\text{ClO}_4$ at ~ 8 K to 145 T. Quadratic magnetoresistance is observed at fields above 100 T. (Bottom) The oscillatory component of the magnetoresistance from the flux compression experiment (solid line), plotted vs. inverse field, compared to results from lower field experiments (60 T) taken in a non-destructive magnet (dashed) at 0.5 K. Good agreement is observed, although the signals are very weak.

oscillations are very weak in both experiments due to the quenched-in disorder of the crystalline lattice. A snubber network (filters) will be added to the capacitor bank on future experiments to help eliminate the switching transient noise.

References:

- ¹ Pioneered by M. Fowler, *et al.*, at LANL.
- ² Brooks, J.S., *et al.*, preprint (1997).

Observation of Both One-Dimensional and Two-Dimensional Angle-Dependent Magnetoresistance Oscillations in $\kappa\text{-(BEDT-TTF)}_2\text{Cu(SCN)}_2$

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The charge-transfer salt $\kappa\text{-(BEDT-TTF)}_2\text{Cu(SCN)}_2$ is perhaps the exemplary organic superconductor. It has been characterized by a wide range of techniques (see e.g. References 1 and 2 and references therein); moreover, the relationship between parameters such as the electronic effective mass and the superconducting properties has been established using pressure,¹ indicating that the superconductivity is probably BCS-like. The Fermi surface is thought to consist of a quasi-two-dimensional hole pocket, separated by a small gap from a pair of open (quasi-one-dimensional) sheets. High-magnetic-field studies³ provide support for this picture via the observation of a high-frequency series of magnetic-quantum oscillations, interpreted as magnetic breakdown between the quasi-two-dimensional and quasi-one-dimensional Fermi surface sections. Recent Hall effect data have challenged this view, however, suggesting that the material enters some form of spin-density-wave (SDW) state at higher temperatures;⁴ if true, this assertion would imply that the low-temperature Fermi surface was very different to the simple prediction described above.

Hall effect data are well known to be open to a wide range of possible interpretations, and so it was decided to check the topology of the low-temperature Fermi surface using angle-dependent magnetoresistance oscillations (AMRO; for a review, see Reference 5). In contrast to magnetic-quantum oscillations, which only give the areas of possible closed semiclassical orbits about the Fermi surface, AMRO enable both closed and open sections of Fermi surface to be

observed, and show in a straightforward manner whether the Fermi surface is reorganized (e.g. by a SDW) or not.⁵

AMRO experiments are only feasible in steady magnetic fields that are well above the superconducting B_{c2} of the material;⁵ in the case of κ -(BEDT-TTF)₂Cu(SCN)₂, this implies that steady fields >20 T are necessary. Therefore the experiments were carried out in 20 MW resistive magnets at Tallahassee, using a special cryostat (built in Oxford) that allows the sample to be rotated through all possible orientations in the magnetic field. AMRO data were recorded at fields of up to 33 T at several temperatures between 1.2 K and 4.2 K.⁶ AMRO due to both open and closed Fermi-surface sections were observed for the first time in this material.⁶ The periods and amplitudes of the AMRO are in good agreement with the proposed Fermi surface⁶ and firmly refute the idea that κ -(BEDT-TTF)₂Cu(SCN)₂ is in some sort of SDW state at low temperatures.

References:

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Magneto Thermoelectric Power and Magnetoresistance of the Doped Polyacetylene

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We have measured the magneto thermoelectric power (TEP) and magnetoresistance (MR) of the

doped polyacetylene ((CH)_x). The anomalous behavior of the magneto TEP near 70 K, i.e. the temperature of abrupt reduction of the magneto TEP for metal halide (AuCl₃, FeCl₃) and iodine doped polyacetylene is investigated as a function of doping concentration. As the doping concentration increases, the reduction temperature of TEP increases. Figure 1 shows the change of TEP, ΔS ($S(T, B=0) - S(T, B=20 \text{ T})$) of (CH(AuCl₄)_y)_x and inset shows the typical magneto TEP for $y=0.071$. Clearly seen from the figure, the reduction temperature of TEP increases from 75 K for $y=0.029$ up to 110 K for $y=0.126$. The reduction of TEP under magnetic field could be due to the spin density wave formation, since the collective motion can have lower entropy than that of the single particle transport. The detailed mechanism is under investigation.

The MR of the fresh sample is negative and independent of the field direction. The magnetic field dependence of MR at low temperature can be fitted with the Kondo-effect like formula, which suggests the importance of the spin-spin interaction between π -electrons in the polymer chain and fixed spins in the dopants located nearby the polymer chain.

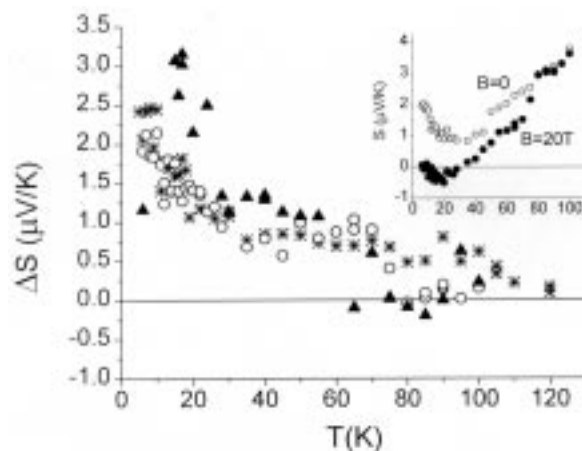


Figure 1. ΔS ($S(T, B=0) - S(T, B=20 \text{ T})$) vs. temperature for (CH(AuCl₄)_y)_x. $y=0.029$ (▲), $y=0.071$ (○), $y=0.121$ (*). Inset shows the magneto TEP results for $y=0.071$.

References:

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Fermi-Surface Study of (BEDO-TTF)₂Cl·3H₂O

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For the quasi-2D conductor (BEDO-TTF)₂Cl·3H₂O as yet unexplained beats in the Shubnikov-de Haas traces (SdH) were reported with the magnetic field slightly misaligned from the normal to the conducting plane. We have used dysprosium flux concentrators to investigate the magnetoresistance to magnetic fields of 36 T and at subkelvin temperatures.¹ We were successful in observing clear beats with a complicated pattern in the field range above 15 T, but the crystal failed to reproduce these features after it was remounted on a two-axis rotating probe. The angle-dependent magnetoresistance oscillations (AMRO) recorded for several different samples show that the material is monoclinic, and the SdH and AMRO data have allowed to reconstruct the most important features of the Fermi surface of the material. Future studies of the material with *in situ* reorientation should allow to correlate the appearance of the beats with the details of the Fermi surface.

References:

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In-Plane Angular Dependent Magnetoresistance Studies of Low-Dimensional Fermi Surfaces—Discrimination of Closed and Open Orbit Contributions

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We have carried out systematic measurements of the angular dependent magnetoresistance (AMRO) on the organic quasi-two dimensional compounds α -(BEDT-TTF)₂KHg(SCN)₄ and α -(BEDT-TTF)₂NH₄Hg(SCN)₄ in magnetic fields up to 33 T. This was accomplished via four terminal AC transport measurements with the current direction along the c axis (perpendicular to conducting planes). AMRO measurements are used to reveal detailed information on materials with a well defined Fermi surface. The Fermi surface topology of these materials is characterized by both 2-d closed cylindrical orbit and 1-d open sheet orbit surfaces. For magnetic field applied parallel to the conducting planes, we find considerable angular dependent anisotropy in the AMRO signal. The magnetic field in this orientation corresponds to values of AMRO taken at theta equal 90 degrees

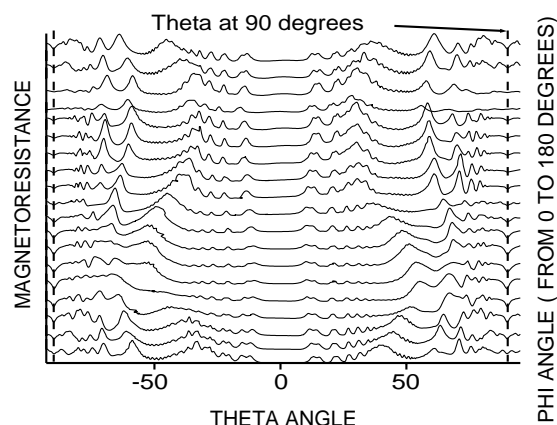


Figure 1. AMRO (Angular dependent magnetoresistance oscillations) for α -(BEDT-TTF)₂KHg(SCN)₄ at 31 T and 1.5 K.

(see Figure 1). We ascribe this anisotropy to the warped nature of the closed and open orbit Fermi surfaces in relation to the magnetic field direction. By comparison with similar studies on materials with only closed orbit Fermi surface sections, we can identify the signature of the open orbit band in the measurements. Our conclusions will be drawn in light of a simple semiclassical Boltzmann transport treatment of the problem. This approach provides new information about low-dimensional Fermi surfaces not accessible by standard (de Haas van Alphen or Shubnikov-de Haas) methods.

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(TMTSF)₂PF₆ in the Extreme Quantum Limit

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The Bechgaard salt (TMTSF)₂PF₆ is an organic quasi 1D conductor that becomes superconducting at temperatures below 2 K when subjected to hydrostatic pressures between 6 kbar and 15 kbar. With the application of magnetic fields the material enters a series of field-induced spin density wave states that leaves the material in an insulating state at the extreme quantum limit. Theoretically it has been predicted that at high enough fields these reduced dimensional systems may undergo a reentrant metallic transition and possibly even go superconducting. To date, experiments have been limited to 33 T due to the lack of a high pressure cell that performed well in the higher fields that can only be achieved with pulsed magnets.

We have performed the first pulsed high magnetic field transport studies of (TMTSF)₂PF₆ at pressures of 8 kbar and temperatures as low as 350 mK. To accomplish this and eliminate eddy current heating that would be seen if a typical metallic high pressure

cell were used, a non-metallic diamond anvil cell was developed.¹ This allows investigation of transport properties well into the extreme quantum limit regime for the first time. Low pressure measurements that put the sample in an insulating state at low temperatures were first performed to verify that there is no eddy current heating using this new cell design. After confirming this, transport measurements were made along the c-axis of the material, which provided a stronger signal, eliminated problems with contact resistance, and allowed for a minimal open loop induced voltage pickup. Figure 1 shows the low field data and the quantum Hall plateaus that have been observed previously.² Data to 60 T (Figure 2) shows the well known rise in resistance at the

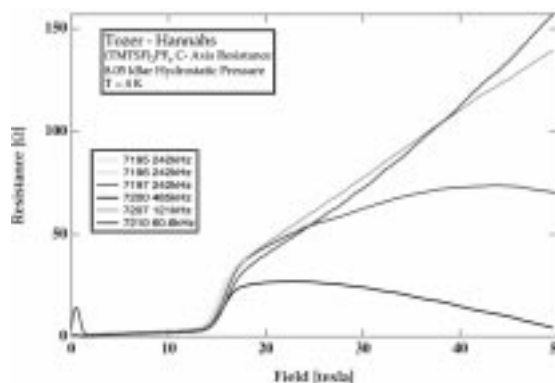


Figure 1. Low field magnetoresistance clearly shows the quantum Hall plateaus in (TMTSF)₂PF₆ at a pressure of 0.8 GPa. Measurements were made at a temperature of 350 mK.

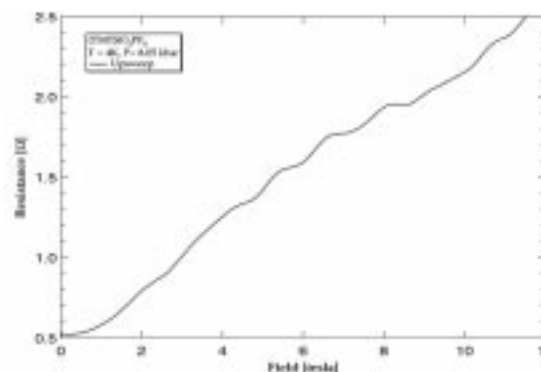


Figure 2. Magnetoresistance of (TMTSF)₂PF₆ showing the clear rise in resistance at $n = 0$. Measurements were made at 0.8 GPa and temperature of 350 mK. The origin of the frequency and field dependence at higher fields has not been ascertained, but may be intrinsic or an experimental artifact. Shots at 60 T closely track this data and show no additional anomalies.

extreme quantum limit. Above this limit the transport shows a marked frequency and field dependence between 80 kHz to 500 kHz and 25 T to 60 T, with the resistance dropping at higher frequencies and fields. It has not been ascertained as to whether this drop in resistance is an experimental artifact or is truly a new transition in this fascinating material. Experiments to 80 T are planned for the near future.

References:

- ¹ The plastic cell is 9 mm in diameter and 12 mm long. With this cell, optical measurements are currently limited to 8.5 GPa. Electrical transport measurements are held to 1.5 GPa at millikelvin temperature due to the gasket design.
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